

Observation of Resistively Detected Hole Spin Resonance and Zero-field Pseudo-spin Splitting in Epitaxial Graphene

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Electronic carriers in graphene show a high carrier mobility at room temperature. Thus, this system is widely viewed as a potential future charge-based high-speed electronic-material to complement- or replace- silicon. At the same time, the spin properties of graphene have suggested improved capability for spin-based electronics or spintronics, and spin-based quantum computing. As a result, the detection, characterization, and transport of spin have become topics of interest in graphene. Here we report a microwave photo-excited transport study of monolayer and tri-layer graphene that reveals an unexpectedly strong microwave-induced electrical-response and dual microwave-induced resonances in the dc-resistance. The results suggest the resistive detection of spin resonance, and provide a measurement of the g-factor, the spin relaxation time, and the sub-lattice degeneracy-splitting at zero-magnetic-field.

The quantum mechanical spin degree of freedom finds remarkable applications in the areas of quantum computing (QC) and spin-based electronics (spintronics).[1–8] For example, in QC scenarios, particle spin often serves as a quantum bit or qubit.[1–6] In spintronics, the spin serves to endow electronic devices with new functionality as in the giant-magneto-resistive (GMR) read-head or the spin-based transistor.[7, 8] Graphene is a novel two-dimensional system with remarkable properties such as massless Dirac fermions, an anomalous Berry’s Phase, a pseudo-spin (valley-degeneracy) in addition to spin, and half-integral quantum Hall effect.[9–12] Graphene is also an appealing material for electron-spin QC and spintronics,[1, 4–8, 13–15] due to the expected weak spin-orbit interaction, and the scarcity of nuclear spin in natural carbon. Because of QC and spintronics, the microwave control and electrical detection of spin have become topics of interest, now in graphene nanostructures,[1–17] where the small number of spins limits the utility of traditional spin resonance.

Here, we report the first observation of resistive detection of spin resonance in epitaxial graphene,[9, 18, 19] provide a measurement of the g-factor and the spin relaxation time, and determine the pseudo-spin (valley - degeneracy)-splitting at zero-magnetic-field. Such resistive resonance detection can potentially serve to directly characterize the spin properties of Dirac fermions, and also help to determine- and tune- the valley degeneracy splitting for spin based QC. [15]

RESULTS

Trilayer graphene. Figure 1(a) exhibits the diagonal resistance, R_{xx} , vs. the magnetic field, B , for the trilayer epitaxial graphene specimen, sample 1. The blue curve obtained at $T = 1.5K$ in sample 1 exhibits a cusp

in R_{xx} near null magnetic field, i.e., Weak Localization (WL),[20–22] followed by positive magneto-resistance at $B > 0.2T$. In Fig. 1(a), an increase in T , to $T = 90K$, results in the red curve, which includes a positive displacement of the R_{xx} vs. B trace with respect to the $T = 1.5K$ trace, i.e., $dR_{xx}/dT > 0$ at $B = 0$ Tesla, along with the quenching of WL. Since WL cannot be observed without inter-valley scattering in monolayer or bilayer graphene,[22] the observed WL is presumed to be an indicator of a non-zero inter-valley matrix-element.

Fig. 1(b) illustrates the influence of microwave-excitation on sample 1 at $F = 48$ GHz. Here, for $B < 1$ Tesla, microwave-excitation produces a positive displacement of the photo-excited R_{xx} relative to the blue trace obtained in the absence of photo-excitation, akin to increasing the temperature, cf. Fig 1(a) and Fig. 1(b). However, at $B > 1$ Tesla, R_{xx} exhibits resistance valleys as the photo-excited curve approaches the dark curve, similar to reducing the temperature. To highlight associated resonances, the change in the diagonal resistance, $\Delta R_{xx} = R_{xx}(4mW) - R_{xx}(dark)$, is exhibited vs. B in Fig. 1(c). Fig. 1(c) shows two noteworthy features: a high magnetic field resonance at $|B| = 1.75$ Tesla, and a low magnetic field feature at $|B| = 1.4$ Tesla. These resonances disappeared upon increasing the bath temperature to $T > 5K$.

Monolayer graphene. Figure 2(a)-(c) exhibit the results for sample 2, while figures 2(d)-(f) show representative data for sample 3. Both sample 2 and sample 3 are monolayer epitaxial graphene specimens. The T-dependence of R_{xx} at $B = 0$ Tesla is shown in Figure 2(a),(d) for samples 2 and 3, respectively. Unlike sample 1, samples 2 and 3 show a decrease in $R_{xx}(B = 0)$ with increasing temperature, i.e., $dR_{xx}/dT \leq 0$. Further, as indicated in Fig. 2(b),(e), microwave irradiation of these specimens produces a uniform negative displacement in the R_{xx} traces with increasing power. Yet, the

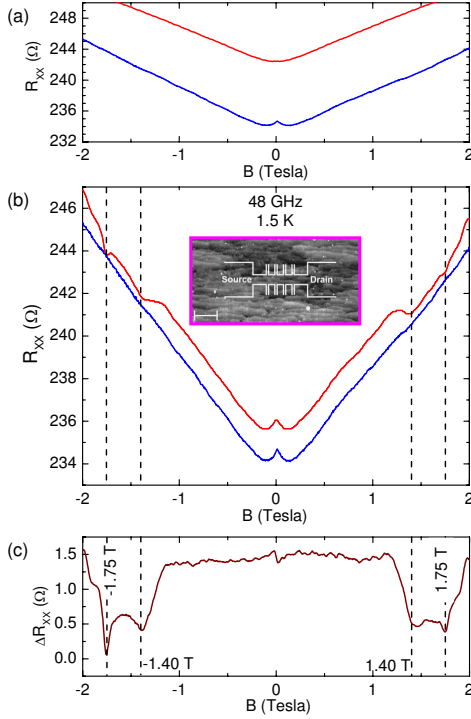


FIG. 1: (color online) **Transport in trilayer graphene** (a) The diagonal resistance, R_{xx} , is shown vs. the magnetic field, B , at temperatures $T = 90\text{K}$, shown in red, and $T = 1.5\text{ K}$, shown in blue, for sample 1, in the dark condition, without microwave excitation. The upward displacement of the $T = 90\text{ K}$ curve with respect to the $T = 1.5\text{ K}$ curve shows that R_{xx} increases with the temperature, i.e., $dR_{xx}/dT \geq 0$. (b) R_{xx} versus B in the absence of microwave excitation is exhibited in blue, and under constant $F = 48\text{ GHz}$ microwave excitation at $P = 4\text{ mW}$ is shown in red, for sample 1. The photo-excited R_{xx} trace shown in red exhibits a uniform upward shift with respect to the dark R_{xx} curve shown in blue for $B < 1\text{ T}$. At higher B , resonant reductions in the R_{xx} are observed in the vicinity of $B = \pm 1.4\text{ T}$ and $B = \pm 1.75\text{ T}$, where the photoexcited R_{xx} approaches the dark value. Inset: An atomic force microscopy image of the EG/SiC surface with the device superimposed upon it. The size scale bar corresponds to $10\text{ }\mu\text{m}$. (c) The change in the diagonal resistance, ΔR_{xx} , between the photo-excited and dark conditions in panel (b), that is, $\Delta R_{xx} = R_{xx}(4\text{mW}) - R_{xx}(\text{dark})$, is exhibited versus B . Note the valleys in ΔR_{xx} in the vicinity of $B = \pm 1.40\text{ T}$ and $B = \pm 1.75\text{ T}$.

effect of microwave excitation ($dR_{xx}/dP \leq 0$ at $B = 0$) is again similar to heating the specimen ($dR_{xx}/dT \leq 0$), cf. Fig. 2(a),(b) or Fig. 2(d),(e). Thus, the $R_{xx}(B = 0)$ from Fig. 2(b),(e) have been marked as colored disks in Fig. 2(a),(d). Apparently, microwave excitation at $P = 10\text{ mW}$ serves to increase the carrier-temperature up to $T = 32\text{ K}$ in sample 2, and up to $T = 36\text{ K}$ in sample 3. At such higher P , the radiation helps to manifest, in addition, resonant R_{xx} peaks in the vicinity of the dashed lines of Fig. 2(b),(e), unlike in Fig. 1(b),

where valleys characterize the resonances in R_{xx} . Yet, in all three specimens, the photo-excited R_{xx} moves towards the dark curve at resonance. Figs. 2 (c),(f) show resonances at nearly the same $|B|$, at $F = 18\text{ GHz}$, in samples 2 and 3.

Spin resonance evolution. Figure 3 illustrates the frequency-evolution of the ΔR_{xx} resonances for all three specimens. Here, Fig. 3(a)-(f) illustrate the results for sample 1, Fig. 3(f) - (h) exhibit data for sample 2, and Fig. 3(i) - (j) show some results for sample 3. Note the shift of resonances to higher B with increasing F .

Figure 4(a) presents a plot of the microwave frequency, F , vs. the resonance magnetic fields, B , extracted from Fig. 3. Fig. 4(a) shows that the resonance B -values for the three specimens collapse onto two lines: a gold-colored line in Fig. 4(a), which represents the high B -field resonances of Fig. 3, exhibits a linear increase as $F(\text{GHz}) = 27.2B(\text{T})$, with the ordinate-intercept at the origin. Another line shown in magenta in Fig. 4(a), which represents the low- B resonances of Fig. 3, exhibits a linear increase as $F(\text{GHz}) = 10.76 + 26.9B(\text{T})$, with a non-zero intercept, $F_0 = 10.76\text{ GHz}$. In such a plot, spin resonance for an electron with g-factor $g_e = 2.0023$ would follow: $F(\text{GHz}) = 28.01B(\text{T})$. Thus, the observed slopes, $dF/dB = 26.9 \pm 0.4\text{ GHz T}^{-1}$ ($dF/dB = 27.2 \pm 0.2\text{ GHz T}^{-1}$) for the low (high) field resonance correspond to spin resonances with $g_{\parallel} = 1.92 \pm 0.028$ ($g_{\parallel} = 1.94 \pm 0.014$).

DISCUSSION

The g-factors measured here are comparable to the g-values obtained from traditional ESR-studies of graphite, which have indicated that the g-factor for $B//c$ -axis, g_{\parallel} , increases from 2.05 at 300 K to 2.15 at 77 K, while, at $T = 300\text{ K}$, the g-factor for $B \perp c$ -axis, $g_{\perp} = 2.003$. [23, 24] In graphite, the g-factor depends upon the orientation of the B field, the temperature, the location of the Fermi level, and the sign of the charge carriers, with opposite g-factor shifts, Δg , from g_e , for electrons and holes. [23–25] The negative Δg and reduced g_{\parallel} observed here relative to g_e are consistent with expectation for holes.

From these data, we also estimate a resonance half-width $\Delta B \approx 0.05\text{ Tesla}$, which corresponds to a spin relaxation time $\tau_s = h/(4\pi\Delta E) = 6 \times 10^{-11}\text{ s}$. The spin relaxation time has been a topic of great interest in graphene. Spin relaxation [26] has been experimentally studied in monolayer and bilayer exfoliated graphene on SiO_2/Si [27–34] and, more recently, on epitaxial graphene [35, 36] using spin valve devices, and possible mechanisms involved in spin relaxation have been examined by theory. [37–39] For monolayer exfoliated graphene, observed spin relaxation times generally fall in the range of 40 - 150 ps (refs [28, 30, 31, 34]) while exfoliated bilayer graphene exhibits spin relaxation

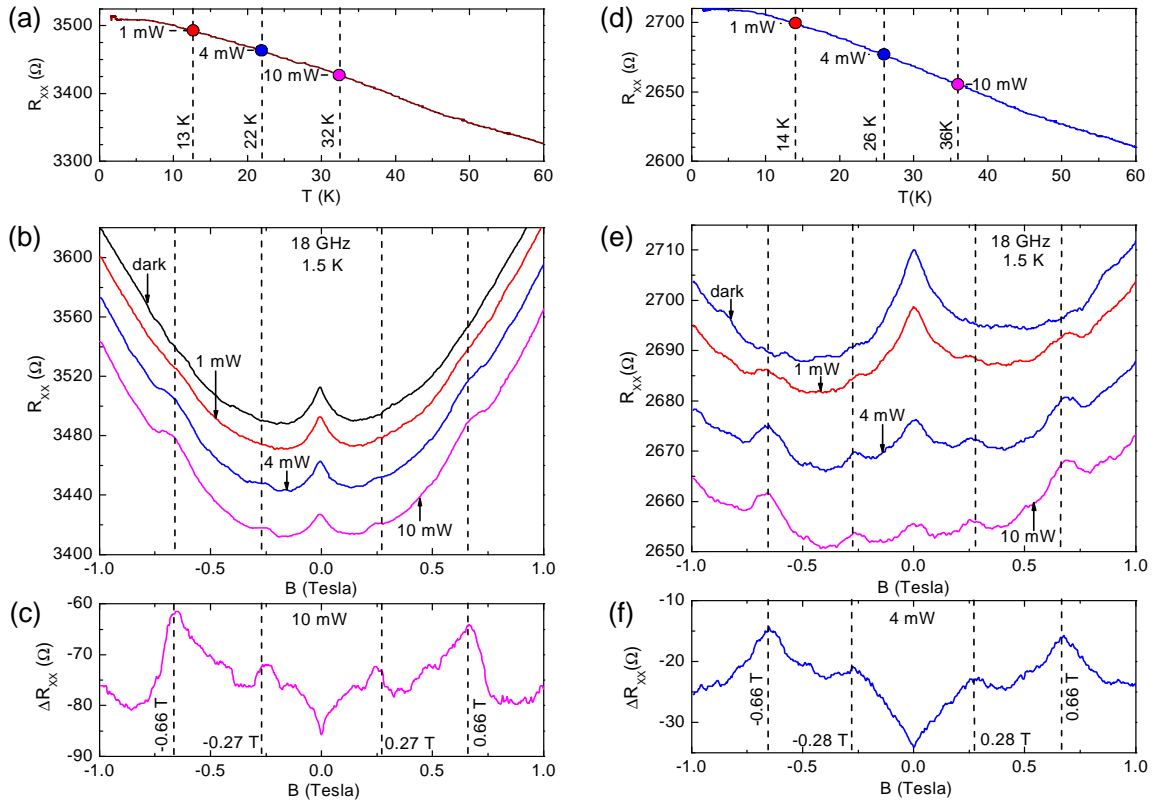


FIG. 2: (color online) **Transport in single layer graphene** Panels (a)-(c) show some results for sample 2, while figures (d), (e) and (f) show representative data for sample 3. (a) Sample 2 shows a decrease in the resistance R_{xx} with increasing temperature T , i.e., $dR_{xx}/dT \leq 0$, in the absence of a magnetic field, i.e., $B = 0$ T. (b) The data traces exhibit a downward shift with increasing microwave power, P , at $F = 18$ GHz, suggestive of microwave induced carrier heating in the specimen. The R_{xx} at $B = 0$ T obtained from these traces have been marked as filled circles in panel (a) above, which shows the T -dependence of R_{xx} . Apparently, $P = 10$ mW serves to increase the carrier temperature up to $T = 32$ K in sample 2. Microwave induced resonances appear in the vicinity of the dashed lines with increasing P . (c) $\Delta R_{xx} = R_{xx}(10\text{mW}) - R_{xx}(\text{dark})$ is shown vs. the magnetic field for sample 2. Note the change in ΔR_{xx} due to spin-resonance in the vicinity of the dashed lines at $B = \pm 0.66$ T and $B = \pm 0.27$ T. (d) Sample 3 also exhibits a decrease in the resistance R_{xx} with increasing temperature T , i.e., $dR_{xx}/dT \leq 0$, in the absence of a magnetic field, i.e., $B = 0$ T. (e) At $F = 18$ GHz and $T = 1.5$ K, R_{xx} is shown versus B for sample 3, at several power levels. The R_{xx} at $B = 0$ T observed in these data have been marked as filled circles in panel (d). (f) This panel shows $\Delta R_{xx} = R_{xx}(10\text{mW}) - R_{xx}(\text{dark})$ versus the magnetic field for sample 3. Note the change in ΔR_{xx} due to spin-resonance in the vicinity of $B = \pm 0.66$ T and $B = \pm 0.28$ T.

times as long as 2 - 6 ns (refs [32, 33]). The observed shorter-than-expected spin lifetime in exfoliated monolayer graphene has been attributed to extrinsic mechanisms based on impurity adatoms,[37] charged impurities and phonons from the substrate,[38] spin orbit coupling due to ripples in graphene,[39] and so on. Note that the τ_s reported here for C -face epitaxial graphene is comparable to previous reports of the spin relaxation time for monolayer exfoliated graphene on SiO_2/Si . The spin-diffusion length here is $\lambda_s = (D\tau_s)^{1/2} = 1.4\mu\text{m}$, [13] while the Hall bar width, $w = 4\mu\text{m}$. Here, D is the diffusion constant. In such a situation, edges could be playing a role in spin relaxation, given that edges in graphene can be magnetically active,[12] and the electrical contacts include gold, a heavy element. Thus, there could be additional av-

enues for spin relaxation in the small specimen, in addition to the other above-mentioned mechanisms.[37–39] Finally, inhomogeneities could serve to broaden the resonance linewidth and help to produce an apparently reduced spin relaxation time.

The observation of similar double resonances in monolayer- and trilayer- graphene can be viewed as a consequence of rotational (non-AB) layer stacking in epitaxial graphene, which makes it possible even for multilayer EG to exhibit the same electronic properties as isolated graphene.[18] Note also that sub-lattice or pseudo-spin degeneracy lifting is known to occur at high- B in graphene.[40–42] For example, the progression of quantum Hall effect from the $R_{xy} = [4(N + 1/2)]^{-1} h/e^2$ sequence,[10, 11, 43] to observations of σ_{xy} increases

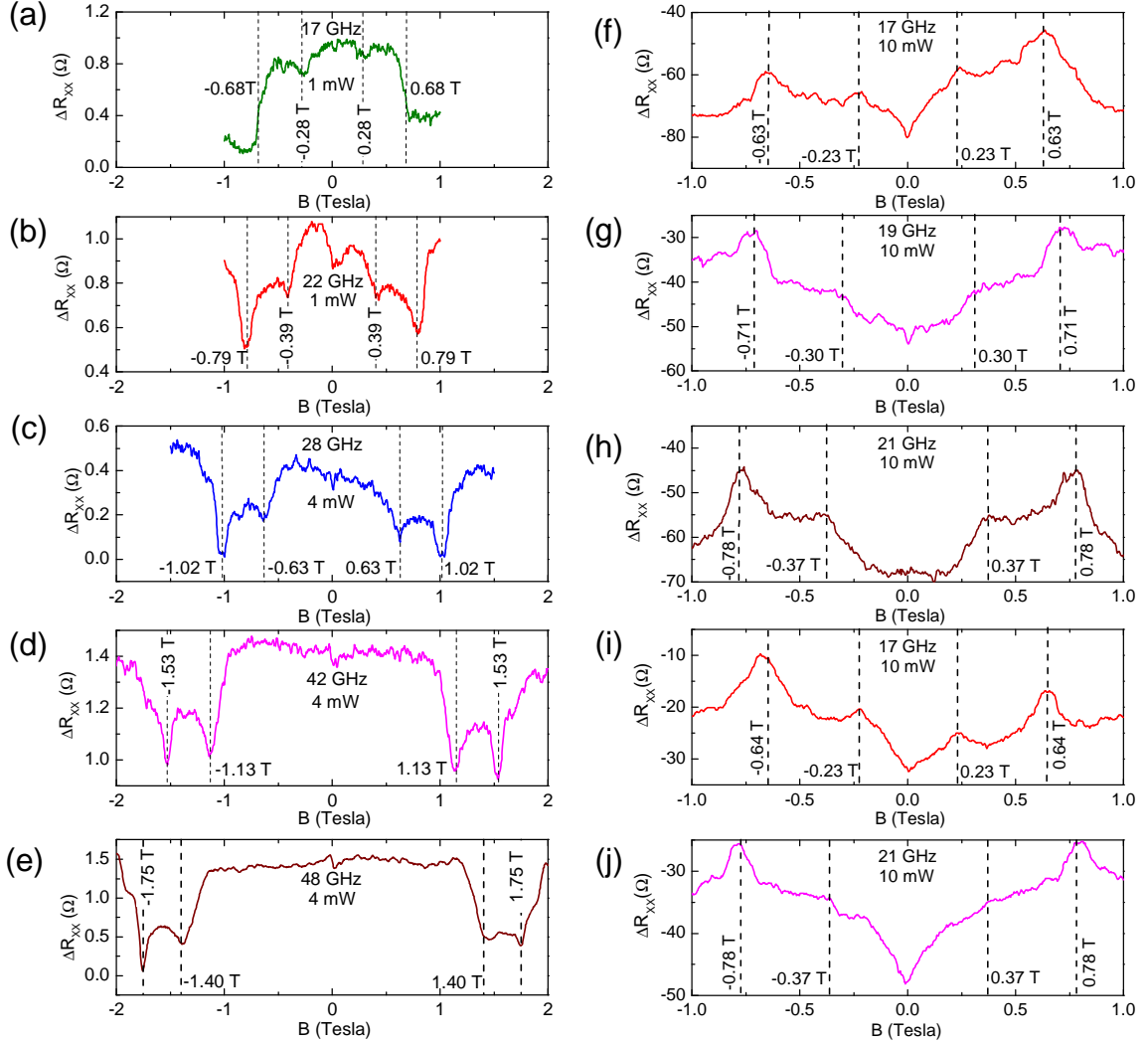


FIG. 3: (color online) **Frequency evolution of microwave-induced resonances** (a) - (e): For sample 1, the photo-induced change in the diagonal resistance, ΔR_{xx} , is plotted vs. B for (a) 17 GHz, (b) 22 GHz, (c) 28 GHz, (d) 42 GHz, and (e) 48 GHz. Note the uniform shift in the resonances, indicated by dashed lines, to higher B with increasing F . Here, the resonances are characterized by ΔR_{xx} minima. (f)-(h): For sample 2, the photo-induced change in the diagonal resistance, ΔR_{xx} , is plotted vs. B for (f) 17 GHz, (g) 19 GHz, and (h) 21 GHz. Note the shift in the resonances, indicated by dashed lines, to higher B with increasing F . Here, the resonances are characterized by resistance maxima. (i)-(j): For sample 3, the photo-induced change in the diagonal resistance, ΔR_{xx} , is plotted versus B for (i) 17 GHz and (j) 21 GHz. Note the shift in the resonances, indicated by dashed lines, to higher B with increasing F .

in steps of e^2/h (ref [40]) reflects the lifting of both the spin- and pseudo-spin- degeneracy. In addition, a non-linear interaction-enhanced valley-degeneracy splitting has been reported from a scanning tunneling spectroscopy study.[42] Finally, the manifestation of weak localization, which is observable in Fig. 1 and 2, is an indicator of inter-valley coupling in these specimens.[22] Since sub-lattice degeneracy splitting is not unexpected due to the above, the observed $F_0 = 10.76$ GHz is attributed to a zero-magnetic-field pseudo-spin (sub-lattice degeneracy) splitting of $\Delta_0 = \hbar F_0 = 44.4\mu\text{eV}$.

A provisional interpretation of the F vs. B plot of Fig. 4(a) is provided in Fig. 4(b). Chiral eigenstates

and linear energy-wavevector dispersion characterize carriers in graphene. The application of a B-field nominally produces fourfold, valley- and spin- degenerate Landau-levels characterized by $E_N = \pm v_F(2\hbar BN)^{1/2}$, where $N = 0, 1, 2, \dots$, e is the electron charge, v_F is the Fermi velocity, and \hbar is the reduced Planck's constant.

We imagine the four-fold degeneracy being lifted by $\hbar F_0$ even at $B = 0$, to produce energy doublets as $E_{N'} = E_N \pm \hbar F_0/2$. Then, owing to the Zeeman effect, associated Landau levels exhibit a further splitting of the spin-degeneracy as $E_{N''} = E_{N'} \pm g\mu B/2$. Observed microwave-induced transitions occur within the highest occupied Landau level in the vicinity of the Fermi level.

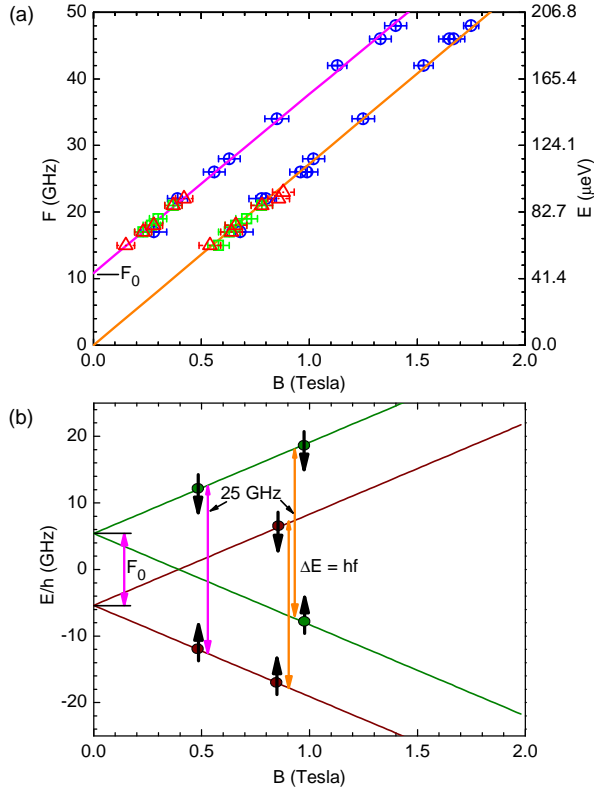


FIG. 4: (color online) **Relation between the resonance magnetic fields and the microwave frequency or energy** (a) This plot shows that the high B -field resonance found in Fig. 3 follows a linear fit, indicated by the gold line, as $F(\text{GHz}) = 27.2B(\text{T})$ with an intercept at the origin. The low B -field resonance found in Fig. 3, indicated by the magenta line, follows a linear fit as $F(\text{GHz}) = 10.76 + 26.9B(\text{T})$ with a non-zero intercept, $F_0 = 10.8$ GHz. Here, the lines indicate the fits, while the symbols exhibit the data points. The circles correspond to the data for sample 1, the squares correspond to the data for sample 2, and the triangles correspond to the data for sample 3. (b) The observed experimental results appear to be consistent with spin-resonance and zero-field pseudo-spin (valley-degeneracy) splitting enhanced spin resonance. A four-fold degeneracy is lifted in the absence of a magnetic field to produce a pair of spin degenerate levels (doublets) separated by $E/h = F_0$. Zeeman splitting then lifts the spin-degeneracy of the upper and lower doublets. Microwave photo-excitation induces spin-flip transitions between the spin-levels of the lower- or upper doublet, as shown by the gold lines. Such transitions require vanishing photon energy in the $B \rightarrow 0$ limit. On the other hand, the transition shown in magenta requires non-vanishing photon energy in the limit of $B \rightarrow 0$.

As $E_N \gg hF_0/2$ and $g\mu B/2$, we remove the E_N term and plot $E/h = (E_{N''} - E_N)/h$ in Fig. 4(b).

Here, microwave photo-excitation induces spin-flip transitions, shown in gold, of unpaired carriers between the spin-levels of the lower or the upper doublet. Such transitions require vanishing photon energy in the limit of

vanishing B . In contrast, a transition between the lower spin ("up") level of the lower doublet and the higher spin ("down") level of the upper doublet requires additional energy hF_0 , and such a transition, shown in magenta, exhibits non-vanishing photon energy in the $B \rightarrow 0$ limit. Thus, the F vs. B plot appears consistent with spin resonance and a zero-field pseudo-spin (valley-degeneracy) splitting enhanced spin resonance.

In summary, we have realized the resistive detection of spin resonance in EG, provided a measurement of the g -factor and the spin relaxation time, and identified- and measured- a pseudo-spin (valley degeneracy)-splitting in the absence of a magnetic field. Such resistive resonance detection can potentially serve to directly characterize the spin properties of Dirac fermions, and also help to determine- and tune- the valley degeneracy splitting for spin based QC.

METHODS

Graphene Samples. Epitaxial graphene (EG) was realized by the thermal decomposition of insulating 4H silicon carbide (SiC).[18] The EG specimens were characterized by ellipsometry and the extracted layer thickness was converted to the number of layers at the rate of 0.335 nm/layer. The c-face of the EG/SiC chip was processed by e-beam lithography into micron-sized Hall bars with Pd/Au contacts. Measurements are reported here for three Hall bar specimens labeled 1, 2, and 3. Sample 1 is nominally trilayer graphene, while samples 2 and 3 are monolayer graphene. The samples are p-type with a hole concentration, $p \approx 10^{13} \text{cm}^{-2}$, and a carrier mobility $\mu \approx 10^3 \text{cm}^2/\text{Vs}$.

Measurement configuration. Typically, an epitaxial graphene Hall bar specimen was mounted at the end of a long straight section of WR-62 rectangular microwave waveguide. The waveguide with sample was inserted into the bore of a superconducting solenoid, immersed in pumped liquid Helium, and irradiated with microwaves over the frequency range $10 \leq F \leq 50$ GHz, at a source-power $0.1 \leq P \leq 10$ mW, as in the usual microwave-irradiated transport experiment.[44] Here, the applied external magnetic field was oriented along the solenoid and waveguide axis as a probe coupled antenna launcher excited the Transverse Electric (TE-10) mode in the waveguide. Thus, the microwave electric field was oriented perpendicular to the applied external magnetic field. The microwave magnetic field lines formed closed loops, with components in the transverse and axial directions of the waveguide.

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AUTHOR CONTRIBUTIONS

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ADDITIONAL INFORMATION

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